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BEHAVIOUR OF NUCLEAR FUEL UNDER IRRADIATION

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Nuclear fuel investigation under the conditions of irradiation is one of important problems of modern nuclear metallurgy. This problem is rather complicated, as far as a neutron flow causes in fissionable materials some specific processes. Relative importance of these processes depends both on material properties and on the conditions of irradiation. This report deals with the results of experimental investigation of irradiation effect on properties of some fissionable materials. It considers a problem of uranium-aluminium interaction under irradiation, a problem of uranium intermetallide amorphization and a problem of eutectic matrix (Al + UAl_4) behaviour. The report provides information on the state of uranium compact and, in particular, UO_2 in spent fuel elements of the "Lenin" atomic icebreaker, as well as on change of uranium dioxide structure dispersed in copper matrix. Besides, the report discusses a problem of structure transformations in the U-3 wt.% Mo alloy and a problem concerning a connection between processes of radiation relaxation and a creep in uranium.

1. Investigation of uranium compounds in aluminium matrix

Some fuel elements are made of uranium dioxide and aluminium. Aluminium possesses more chemical affinity to oxygen than uranium. That is why there is a possibility of interaction between uranium dioxide and aluminium in the course of manufacturing and irradiation. The works [1,2] show that such an interaction is going on rather quickly at temperatures of 500-620°C. Investigation [3] of compatibility of uranium dioxide and aluminium under irradiation show that

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irradiation during thousands of hours at temperatures of 100-300°C the reaction between UO_2 and Al is also possible. As a result of an interaction a laminated structure was formed in pellets. Inner layers the temperature of which in the initial period, were 300°C, consisted of UAl_4 . Outer layer with a irradiation temperature of about 100°C, consisted of UAl_3 and UO_2 . A compound UAl_2 was formed in outer layers of some pellets.

Table 1 gives data on interaction between UO_2 and Al. Interaction reaction between uranium dioxide and aluminium results in formation of various intermetallides UAl_2 , UAl_3 and UAl_4 and in volume reduction. But with the presence of adsorbed gas and water in uranium dioxide and aluminium powders the reaction progresses with a volume increase that in some cases results in damage of the fuel element (Fig.1). As far as an interaction between uranium dioxide and aluminium takes place, it is unlikely that the use of uranium dioxides as a nuclear fuel in elements with aluminium matrix will be expedient. Along side with UO_2 in aluminium, was also studied the behaviour of compounds $\text{UAl}_3 + \text{UAl}_4$ in eutectic matrix of alloy containing 23 wt. per cent of U. An X-ray analysis of the alloy revealed beside aluminium two other phases corresponding to compounds UAl_4 and UAl_3 . Measurement of micro-hardness also shows the presence in the alloy of two uranium phases with a hardness $H_V = 250 \text{ kg/mm}^2$ and $H_V = 480 \text{ kg/mm}^2$ and an eutectic with a hardness $H_V = 60 \text{ kg/mm}^2$.

The 90 per cent enrichment of uranium was used in the alloy. Experimental alloy samples in aluminium cans were irradiated at a surface temperature of 120°C up to the 25 and 53 per cent burn-up of uranium-235.

After irradiation the samples were in a satisfactory state. A metallographical analysis of the irradiated alloy revealed two structure components: a uranium intermetallide and an eutectic. There is a ring around the intermetallic uranium grains. The ring copies a configuration of grains (Fig.2). The ring width is 10μ that probably corresponds to the fission fragment path length in the eutectic (Al+ UAl_4).

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Microhardness of the grain is $H_V = 325 \text{ kg/mm}^2$, that of the ring - $H_V = 265 \text{ kg/mm}^2$ and the matrix microhardness decreases from the grain boundary toward periphery from 200 down to $150\text{-}135 \text{ kg/mm}^2$.

It can be supposed that the revealed ring is a matrix region subjected to an additional effect of fission fragments.

Calculations prove that while a mean concentration of fission fragments, after the 48 at per cent burn-up of uranium, are within the eutectic matrix $\bar{c} = 1.5$ per cent, the ring concentration reaches $\bar{c} = 5.0$ per cent. Measured microhardness of the matrix with different concentration of fission fragments is presented in Fig.3.

An X-ray analysis of irradiated samples reveals only aluminium lines shifted toward small angles. Uranium intermetallide lines are not observed in X-ray patterns, though in photomicrographs the intermetallide crystals can be distinctly seen (Fig.2).

Parameter of aluminium lattice in U-UAl_4 eutectic after irradiation up to the 23 per cent burn-up of uranium atoms increased from $a_0 = 4.041 \pm 0.001 \text{ kX}$ up to $a = 4.048 \pm 0.02 \text{ kX}$ and after the 48 per cent burn-up - to $a = 4.051 \pm 0.002 \text{ kX}$ that caused a volumetric swelling of aluminium up to .75 per cent (Fig.3).

An increase of aluminium lattice parameter is caused by uranium burn-up and connected with incorporation of fission fragments into aluminium lattice. Disappearance of intermetallide lines was probably caused by a structure amorphization.

2. Study of compact uranium dioxide

A study was conducted to observe the behaviour of uranium dioxide compact under irradiation at different power per length unit. Samples in the shape of pellets with a diameter from 4 to 40 mm were prepared for that studying.

U-235 enrichment was predetermined in accordance with the power needed and varied within the range .7 - 90 per cent. Uranium dioxide density was 75-95 per cent of the theoretical one. Samples were irradiated under a powers varied from a few w/cm up to 750 w/cm.

Uranium dioxide in fuel elements of the "Lenin" icebreaker was irradiated at a power $q_0 = 750$ w/cm. Irradiation with such a power could not cause a core center melting [4]. True, the only cracking of uranium dioxide pellets can be observed after their irradiation (Fig.4). Distribution of cracks across the pellet section is quite irregular.

Pellet cracking was probably caused by an effect of tensile thermal stress. The value of the latter at a temperature drop across the pellet section $\Delta t = 530^\circ\text{C}$, reaches $\sigma = 33$ kg/mm², that two times exceeds the strength limit of UO_2 within a temperature range of $20-1,000^\circ\text{C}$ [5]. Restraining action of the can becomes effective only for a gap between uranium dioxide pellets and can $\delta = .116$ mm, i.e. rather less than actual gap $\delta = .05$ mm.

Occurrence of a cavity in the core center observed in experiments in UO_2 irradiation with power from 290 w/cm and more (Fig.4). A column-shaped structure of uranium dioxide was revealed side by side with the central cavity.

During uranium dioxide short-term irradiation with a high power ($q_0 = 750$ w/cm, $\tau = 4$ hrs) the column-shaped grains are rather porous and the central cavity makes up only .4 of the pellet diameter. An increase of irradiation period under the same conditions results in column-shaped grain densification due to pore diffusion in radial direction toward the central cavity. Cavity size increases from .4d to .63d, i.e. from 1.8 mm to 2.6 mm.

Cavity occurrence in the central zone of uranium dioxide pellets is probably connected not only with a center melting but also with pore diffusion from the outer surface to the center. As it is seen, at sufficient stiffness of the cans structural transformations within compact uranium dioxide are not dangerous.

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3. Change of uranium dioxide lattice parameter during irradiation

Studying was conducted with uranium dioxide which had an initial lattice parameter $a_0 = 5.461 \pm .001 \text{ kX}$ that corresponded to the formula $\text{UO}_{2.05}$. Uranium dioxide was dispersed in a copper matrix. A relative density of uranium dioxide was 90 per cent of the theoretical one. UO_2 particle size chosen was as minimal as possible (negative fraction - 60μ).

A metallographical analysis proved that the contents of fractions with particle sizes more than 20μ and less than 20μ were approximately equal. As the path length of fission fragments in uranium dioxide was in the range of $9-12 \mu$ [6, 7], a rough estimate of the fragment quantity leaving UO_2 gave a value in the range of 50-60 per cent.

A uranium dioxide volumetric content in a pressure-sintered compound $\text{UO}_2\text{-Cu}$ was 32 per cent. Compound porosity did not exceed 4-5 per cent.

Samples were irradiated in the reactor RFT up to burn up of one third of all the uranium atoms. Sample surface temperature was $80-90^\circ\text{C}$. In the center of samples (pellets of 5-mm in diameter) a calculated value of the temperature in the first moment after the reactor starting did not exceed 120°C .

An X-ray diffraction analysis of irradiated uranium dioxide proved that its lattice parameter reduced considerably from $a_0 = 5.46 \text{ kX}$ down to $5.40 \pm .02 \text{ kX}$.

It proceeded from the calculation that the amount of released oxygen during burn-up of one third of uranium atoms is quite sufficient for oxidation up to the composition UO_3 . Nevertheless, according to [8] the lattice parameter $a = 5.40 \text{ kX}$ corresponds to the composition $\text{UO}_{2.25}$. Therefore it can be expected that the rest portion of the released oxygen will diffuse to pores and participate in fission fragment oxidation.

Thus it was demonstrated that a considerable portion of oxygen releasing at uranium fission is consumed for an ad-

ditional oxidation of uranium without volume increase. Such uranium dioxide behaviour is advantageous when used in metal matrices which do not interact with it.

4. Structure transformations in alloy of uranium with three per cent molybdenum under irradiation

Works [10] and [11] showed that under an irradiation a heterogeneous structure of uranium alloys with 9-10 wt per cent of molybdenum is transformed into a single-phase solid solution. Such a transformation was explained [9] by a process of a specific radiation diffusion due to atom mixing in thermal spike volume. As it is known γ -solid solution in rich molybdenum uranium alloys is very stable at low temperatures and disintegrates only after prolonged exposures within the temperature range of 350-550°C. Therefore, both from the scientific and practical points of view it was worth to study phase transformation caused by irradiation in alloys with a low percentage of molybdenum, metastable solid solutions of which is low stable. Some preliminary experiments in studying an influence of a neutron irradiation on structure changes in an alloy of uranium with three per cent of molybdenum will be considered below.

An alloy containing 96.48 wt. per cent of uranium and 3.32 wt. per cent of molybdenum was taken as a material to be tested. As a result of a high-temperature quenching an δ' -solid solution oversaturated by molybdenum is detected in this alloy at a room temperature. A high-temperature annealing causes a solid solution disintegration into phases δ and U_2Mo accompanied by a reduction of electrical resistance measured at a room temperature on 40 per cent. A disintegration process develops rather quickly (within several hours) in the temperature range of 400-550°C. Disintegration rate reduces with an annealing temperature drop. At temperatures lower than 250°C the solid solution disintegration is not take place practically.

Samples were irradiated in a flow of $2 \cdot 10^{13}$ n/cm²sec at a temperature of $190 \pm 10^\circ C$ and at a temperature lower than

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100°C. Prior to irradiation the samples were in two states. A portion of samples was quenched from 800°C in water and another portion after quenching from 900°C in oil was subjected to annealing at 515°C during 50 hours.

Results of electrical resistance changes of the both sample groups after different periods of irradiation are presented in Table 2. It can be seen from this Table that irradiation of heterogeneous samples resulted in a gradual increase of their electrical resistance which reached 11.6 per cent after a radiation dose of about $6 \cdot 10^{19}$ n/cm². According to value and rate of an electrical resistance change an increase of resistance cannot be account for accumulation of radiation defects only [10], [12]. Therefore it should be supposed that an electrical resistance increase in heterogeneous samples is caused by an alloy radiation homogenization similar to that which was observed in rich molybdenum alloys.

Electrical resistance of quenched samples with a pre-irradiated structure of a single-phase solid solution reduces in the course of irradiation. Decrease of electrical resistance in quenched samples progresses quickly in the course of irradiation at a temperature of 190°C and rather slowly in the course of irradiation at temperatures lower than 100°C. Such a change of electrical resistance can be explained only by the fact that a disintegration of solid solution occurs in the course of irradiation at temperatures mentioned above. As it was already said, the process of a solid solution disintegration in ordinary conditions did not practically occur at temperatures lower than 250°C. Thus it can be concluded that an irradiation considerably activates the disintegration process of oversaturated solid solution in the alloy of uranium with 3 per cent of molybdenum. Evidently, an acceleration of the disintegration process is connected with the fact that irradiation causes an increased concentration of defects in the metal and thus facilitates a diffusion of atoms within short distances. As far as during irradiation at relatively low temperatures the disintegration process of oversaturated solid solution is mainly governed by the diffusion rate, an increase of this rate will cause a considerable acceleration of the disintegration process.

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Thus, the mentioned results testify a quite different behaviour of uranium alloys with 9 and 3 per cent of molybdenum under irradiation. As a quenched state in an alloy of uranium with 9 per cent of molybdenum is very stable and even can be formed in the course of irradiation from a two-phase structure, the α -solid solution, formed as a result of quenching an alloy of uranium with 3 per cent of molybdenum and rather stable in ordinary conditions, under the irradiation quickly disintegrates.

5. Radiation relaxation and creep of α -uranium

The works [10] showed that micro- and macrostress relaxation occurs in samples of α -uranium under the effect of neutron irradiation. The fact that a stress relaxation in α -uranium unlike β -uranium practically continues till zero made it possible to suppose that radiation creep and radiation relaxation in this material are developed with one and the same mechanism. If the in relaxation experiment might be repeated with one sample many times without change of the stress decrease rate in each stage, it should be evident that the radiation relaxation and creep must be identified. In this case the creep rate at the steady stage might be simply equated with relaxation rate at the load which corresponds to the load in the creep experiment. Therefore it was worth to reproduce the stepped relaxation of stresses of the deflection in the α -uranium plates in the following way.

An α -uranium plate spring, $30 \times 1.5 \times 0.1$ mm in size was placed between half-cylinders bending it in the form of an arc with a radius selected so that the highest stress in the spring reaches about 15 kg/mm^2 . Being so elastically bent the spring was subjected to irradiation at a temperature of 80°C in a flow of $2.7 \cdot 10^{12} \text{ n/cm}^2\text{sec}$ and from time to time the irradiation was interrupted to conduct measurements of the deflection. After the first term ($1.4 \cdot 10^{18} \text{ n/cm}^2$) the spring was additionally bent so that the new maximum stress again reached the initial value. Four steps of relaxation

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were obtained in this way (Fig.5) . In Fig.6 these data are represented by semi-logarithmic co-ordinates. As it can be seen from this figure, the relaxation rate in the second and next steps remains practically unchangeable that testifies probable identity of the radiation relaxation and creep processes in the second and next steps. In the beginning of the first stage the relaxation develops with a higher rate than in the next stages. Thus the stress relaxation for

α -uranium can be used yet from the second stage as rather simple method of trial for the creep.

It is of interest to compare our data with the results given in the work by Roberts and Cottrell 13 . A creep rate determined by a formula given in their work, for example, for a stress of 5 kg/mm^2 is $0.57 \cdot 10^{-9} \text{ sec}^{-1}$. Deformation rate determined by our curves makes up $0.47 \cdot 10^{-9} \text{ sec}^{-1}$. Such a good coincidence of data testifies in favour of the conclusion concerning an identity of the radiation relaxation and creep processes.

Table 1

INTERACTION OF UO_2 WITH Al MATRIX IN THE COURSE OF IRRADIATION

Ser. Nos.	Compound	Content of UO_2 in compound UO_2+Al		Enrich- ment , %	Tem- pera- ture, $^{\circ}C$	Time of ir- radia- tion, hrs	U-235 burn- up , %	Atom burn- up , %	Quantity of fis- sioned atoms , $1/cm^3$	Fuel layer swel- ling, %	Composition of nuclear fuel layer according to data of metallo- graph and X-ray ana- lyses
		weight per cent	volumetric per cent								
1	UO_2+Al	46	16.7	20	120	5,400	10	0.14	$8.0 \cdot 10^{19}$	5.5	$UO_2+Al+UAl_x$
2	UO_2+Al	35	12.0	75	120	6,200	50	1.82	$11.2 \cdot 10^{20}$	11.0	$UO_2+Al+UAl_x$
3	UO_2+Al	35	12.0	75	120	11,000	80	2.91	$17.8 \cdot 10^{20}$	17.0	$UO_2+Al+UAl_x$

Electrical resistance changes of a uranium alloy with
3 per cent of molybdenum in the course of irradiation

Ser. Nos.	Irradia- tion tempe- rature, °C	Time of irradia- tion in the flow of $2 \cdot 10^{13}$ n/cm ² sec, hrs	Change of electrical resis- tance $\frac{R}{R}$ %	
			Annealed samp- les	Quenched samp- les
1	190	1	+ 0.9	- 0.9
2	190	3	+ 1.7	- 2.5
3	190	9	+ 2.5	- 9.0
4	190	24	+ 5.6	-12.3
5	100	346	+ 7.5	-11.5
6	100	840	+11.6	-15.0

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Fig 1. Failed UO_2 + Al based fuel element canned in aluminium.

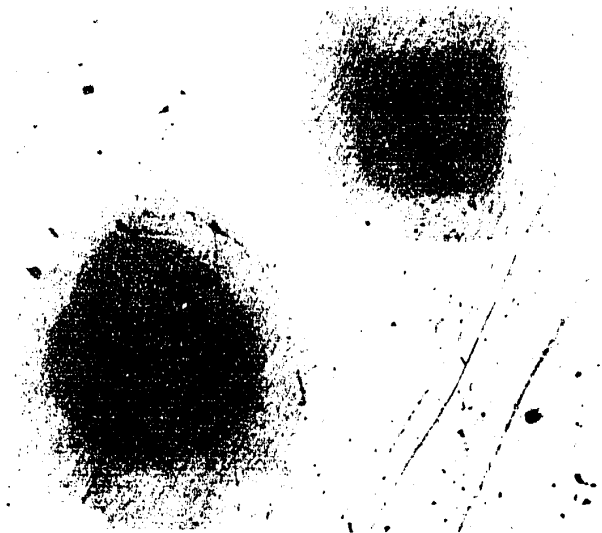


Fig 2. Aluminium-22 w/o uranium alloy structure after irradiation to 48% burn-up in reactor *RFT*. The ring around intermetallic uranium grains is a result of the affect of fission fragment on the matrix.

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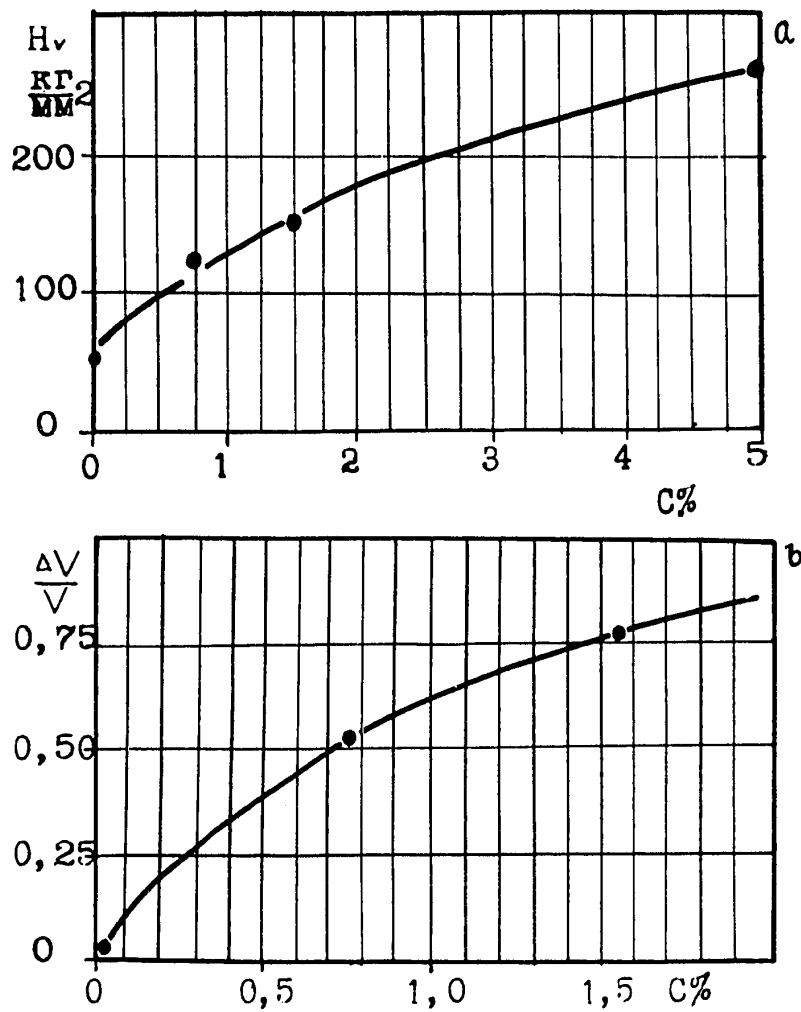


Fig 3. Microhardness of the $Al+UAl_4$ eutectic (a) and the aluminium lattice volume expansion (b) as a function of fission fragment concentration in the aluminium crystal lattice.

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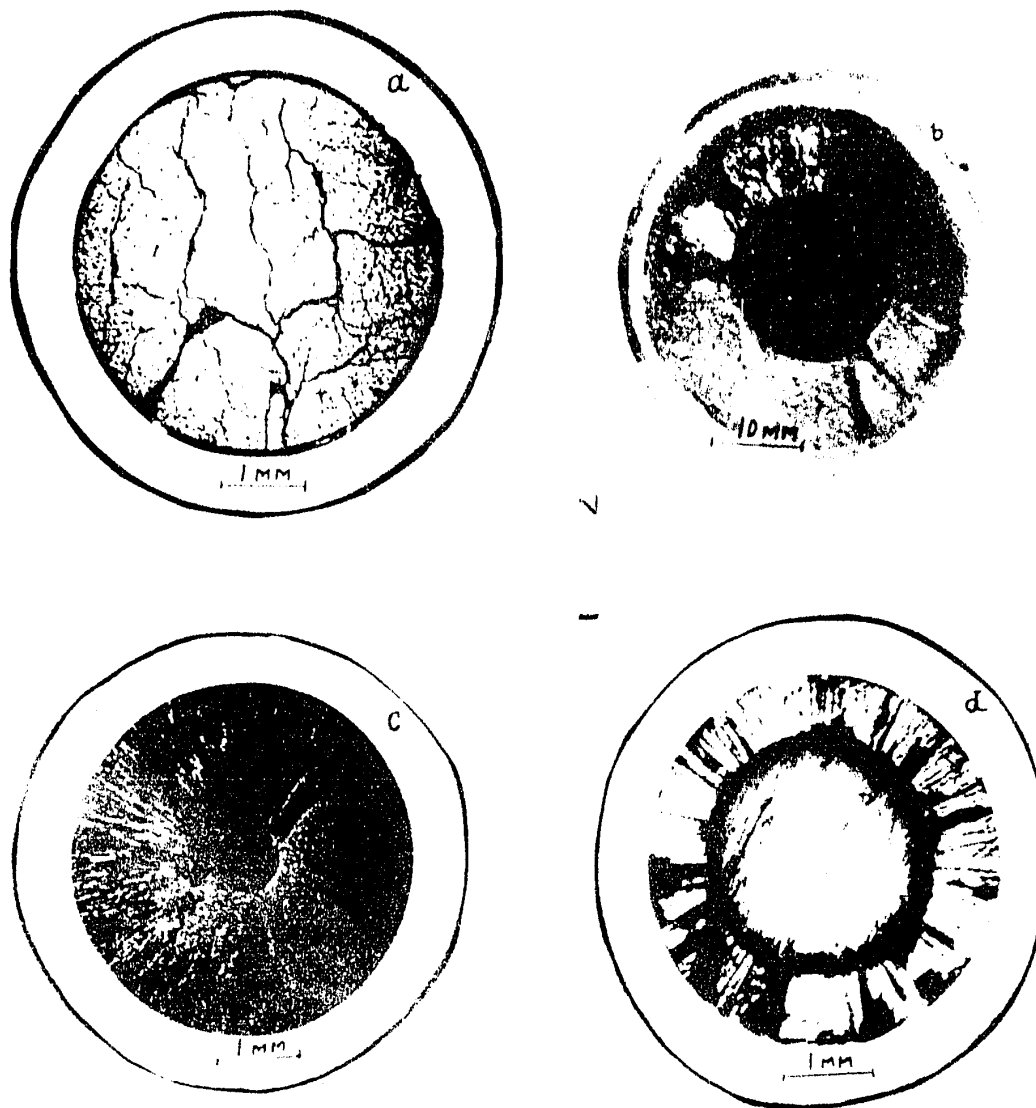


Fig.4. Uranium dioxide after irradiation of different power.

- a) $q_p = 150 \text{ w/cm}$, ice-breaker "Lenin"
- b) $q_p = 290 \text{ w/cm}$,
- c) $q_p = 750 \text{ w/cm}$, $t = 4 \text{ hours}$
- d) $q_p = 750 \text{ w/cm}$, $t = 200 \text{ hours}$

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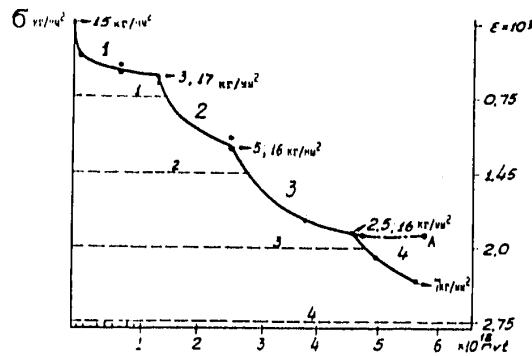


Fig 5. Stepped relaxation. The distance between the dashed (horizontal) line and the origin of the corresponding part of the curve, designated by the same figure, shows the value of the maximum stress at the given step (1,2,3,4) of the relaxation curve (A-relaxation of the irradiated spring on annealing at 100°C without irradiator

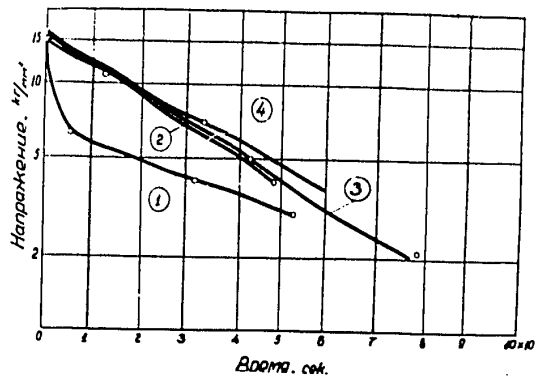


Fig. 6. Variation of stresses at each steps as a function of time of irradiation in the flux of $2.7 \cdot 10^{12} \text{ n/cm}^2 \text{ sec}$. The figures denote the number of step.

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